Questions to TF HTAP for the Review of the Gothenburg Protocol

The review of the Gothenburg Protocol (GP) currently consists of an annotated outline structured into several chapters, each with a different focus. Each chapter consists of several questions assigned to different subsidiary bodies under the LTRAP Convention. Subsidiary bodies have been invited to include additional information in their answers to these questions as appropriate.

TF-HTAP has been assigned to contribute to answers under two different chapters: one chapter with a specific focus on hemispheric transport; and another with a more general focus on measurement and modelling activities contributing to the LRTAP Convention. TF-HTAP will initially focus on providing self-contained answers to all of its assigned questions under the chapter on hemispheric transport.

In this document, the questions assigned to TF-HTAP have been grouped into four topics, around which we plan to structure the GP review chapter focused on hemispheric transport.

Instructions to Readers and Contributors

*Italics*

Notes in italics identify the chapters of the GP review to which topics were assigned, and the subsidiary bodies under the LRTAP Convention that were asked to address each question under those topics. Information from each of the bodies will be integrated to formulate a full answer. Recognizing that other subsidiary bodies may be better positioned to answer aspects of each question, the TF HTAP leadership team has highlighted what aspect of the question we believe the TF HTAP is best suited to address.

- **Bullet Points**
  
  Bullet points are intended to be summary statements that capture the most important points of each answer. The text and citations that follow the bullets are intended to provide further details and references.

**Duplication**

We realize that there is some degree of overlap between these topics and questions. We expect that this duplication will be reduced once all subsidiary bodies of the LRTAP convention have provided their initial input.
Topic 1: Contribution of hemispheric transport to observed trends in air quality and its impacts, and future projections

This topic consists of two questions posed by the WGSR, each assigned to different chapters of the GP review. Question 3.2 was assigned to the hemispheric transport chapter, and question 2.1 was assigned to a chapter on the measurement and modelling activities of the convention. The hemispheric transport chapter of the GP review will begin with an answer to question 3.2 that also answers the elements of question 2.1 concerning hemispheric transport.

Questions under this topic

Question 3.2: What is the current contribution and will be the expected future contribution of emission sources outside the UNECE region to ecosystems and health impacts in the UNECE region, in particular for ozone, PM (and BC)?

This question was assigned to TF HTAP and MSC-W.

Question 2.1:

a) What are the observed and projected trends in air quality for ozone, SO2, PM (species) and oxidised and reduced nitrogen (in the UNECE region)?

b) To what extent are these trends associated with emission trends in the region or dependent on transcontinental transport of air pollutants?

c) What are the observed and projected trends in urban air quality? What is the contribution of long-range transport to air pollutant concentrations in cities? What is the distance to the WHO air quality guideline values (including to updated values, if available on time)?

This question was assigned to MSC-W, TFMM, TF HTAP, TFIAM, and EPCAC. Observed and projected trends at the regional and urban scales in the EMEP region will be addressed best by CCC, TFMM, MSC-W, TFIAM, and EPCAC. Observed and projected trends in North America will be addressed best by the United States and Canada. For purposes of addressing this question, TF HTAP should focus on placing the observed and projected regional trends into the context of observed and predicted trends outside the UNECE region and at the global scale.

Summary of the TF-HTAP contribution to this topic

1. The hemispheric contribution to ground-level ozone is larger than the hemispheric contribution to PM or its components due to ozone’s longer atmospheric lifetime. The concentration of ozone experienced at any given location is the combination of ozone and ozone precursors transported from distant sources on hemispheric to regional scales and, depending on the photochemical regime, local photochemical ozone production or local ozone loss due to titration with NO. Reduction in emissions of ozone precursors in the UNECE region has led to a reduction in peak, short-term ground-level ozone concentrations associated with local photochemical production, especially in the summertime. Reduction of
NO\textsubscript{x} emissions has also led to a reduction in the titration of ozone by NO, leading to higher concentrations of ground-level ozone, especially between autumn and spring, at nighttime, and in Europe. Both effects have increased the relative influence of background ozone, including ozone from hemispheric transport, on local concentrations of ozone experienced in urban areas of the UNECE region, but especially in Europe.

2. Peak ground-level ozone levels in Europe and North America have decreased strongly since 2000, but trends for annual average ozone levels are mixed, with increases at some sites and decreases at others. Average ozone levels in the free troposphere above Europe and North America, as measured by aircraft, have continued to increase. In other parts of the world, both peak levels and annual average levels of ground-level ozone have continued to increase, as have ozone levels aloft as measured by aircraft.

3. The mixed or weak trends in annual average ozone levels belie opposing trends in different seasons. In Europe, in winter (DJF) and spring (MAM) some sites have experienced weak increasing trends and others weak decreases. In summer (JJA), however, most European sites have had strong decreases over the period 2000-2014. In autumn (SON), most sites have seen no trend or a weak decrease. In North America, winter (DJF) ground-level ozone levels strongly increased over the period 2000-2014 and summer (JJA) levels strongly decreased. Trends in spring and autumn were mixed with many sites showing no significant trends. (Chang 2017).

4. This observed trend in ground-level ozone and its impacts cannot be explained completely by precursor emission trends in Europe and North America. Downward trends of ozone precursor emissions in Europe and North America since around 1990 appear to be at least partially offset by increasing NO\textsubscript{x} and VOC emissions outside the UNECE region and increasing CH\textsubscript{4} emissions globally.

5. The contribution of anthropogenic emission sources outside the UNECE region to PM species and their associated impacts within the UNECE region is negligible compared with the impact of local anthropogenic sources. Wildfires and wind-blown dust emanating from outside the UNECE, however, do influence PM levels and deposition in the UNECE region and are sensitive to changes in climate.

6. The absolute contribution of NO\textsubscript{x} and VOC emissions outside the UNECE region to annual average ground-level ozone in Europe and North America is not expected to change significantly under a business as usual scenario to 2050. Expected increases in global CH\textsubscript{4} are expected to more than offset projected reductions of NO\textsubscript{x} and VOC emissions in Europe and at least partially offset reductions of NO\textsubscript{x} and VOC emissions in North America.

7. If NO\textsubscript{x} and VOC emissions were reduced everywhere by the same percentage, the emission reductions outside of Europe would have a bigger impact on European ozone levels than the emission reductions within Europe. In North America, equal percentage emission reductions of NO\textsubscript{x} and VOC outside of North America would contribute significantly to decreases of ozone in North America, but not more than the equal percentage emission reductions in North America itself.
Further explanation of the TF-HTAP contribution to this topic

Observed Trends

Ground-level O$_3$ observations for the period 2000-2014 collected for the global Tropospheric Observation Assessment Report (TOAR) showed that peak O$_3$ values strongly decreased in North America and Europe, and strongly increased in parts of East Asia. However, the trends were more mixed for summer daytime average O$_3$ concentrations in North America and Western Europe, with some sites showing significant increases (Chang 2017; Schultz 2017).

Observations of ozone aloft from commercial aircraft show increasing median annual average ozone levels throughout the troposphere above all 11 regions for which data is available in the period 1994-2016. Maximum ozone levels (95$^{th}$ percentile observations) also increased above most regions, except in the boundary layers of eastern North America and Europe, which experienced decreases (Gaudel 2020).

Parrish et al. (2020) suggest that ozone observations from 6 surface and 5 sonde sites selected to represent the inflow to Europe and North America show very similar seasonal cycles and long-term trends, increasing before 2000, reaching a maximum in the mid-2000s, and decreasing slowly to 2017-2018 (Parrish 2020). Considering ozone observations at a set of 20 remote surface sites in the Northern Hemisphere that overlaps the set considered by Parrish et al. (2020), Cooper et al. (2020) find an even split between increasing and decreasing trends over the period 1995 to 2017-2018. There is uncertainty about the extent to which observed trends at individual surface sites represent changes in hemispheric background or long range transport, as opposed to changes in local or regional sources.

The mixed or weak trends in annual average ozone levels belie opposing trends in different seasons. In Europe, in winter (DJF) and spring (MAM) some sites have experienced weak increasing trends and others weak decreases. In summer (JJA), however, most European sites have had strong decreases over the period 2000-2014. In autumn (SON), most sites have seen no trend or a weak decrease. In North America, winter (DJF) ground-level ozone levels strongly increased over the period 2000-2014 and summer (JJA) levels strongly decreased. Trends in spring and autumn were mixed with many sites showing no significant trends. (Chang 2017). Zhang et al. (2020) estimated that the modeled global tropospheric ozone burden increased by 9% from 1980 to 2010. Half of that increase was due to an increase in global emissions of ozone precursors. The other half of the increase was due to an equatorward shift in emissions, as emissions growth has occurred in the tropics where meteorological conditions are favorable to ozone production.
For fine particles, satellite observations over the period 1992-2012 show decreasing trends in North America and Europe and strong increasing trends in South and East Asia (Boys 2014). Analyses of more recent satellite observations have shown decreasing emissions of NO\textsubscript{X} and SO\textsubscript{2} and decreasing PM over East Asia, which have been attributed to the implementation of emissions control policies (Karplus 2018; Liu 2017; Zheng 2018).

**Emissions Trends**

Between 1990 and 2010, anthropogenic emissions of PM increased little globally, but shifted geographically, with a 30% decline in Europe and North America and a 50% increase in Asia (Klimont 2017). Similarly, between 2000 and 2010, global anthropogenic emissions of ozone precursors (NO\textsubscript{X}, CO, and NMVOCs) grew modestly. However, emissions in Europe and North America decreased by 10% to 50% while emissions in South Asia and East Asia and other regions of the world increased by 10% to 50% (Turnock 2018). Globally, anthropogenic CH\textsubscript{4} emissions increased by 17% between 1990 and 2012, with decreases in Europe, little change in North America, and strong increases in East Asia, South Asia, and other regions of the world.
Since 2010, there have been strong decreases in SO2 emissions and significant decreases in NOx, CO, and PM emissions in China. NMVOC emissions have continued to increase outside of Europe and North America, particularly in Africa and Asia. Likewise, NH3 emissions have continued to increase outside of Europe and North America, particularly in East and South Asia (McDuffie 2020).

The changing spatial patterns of emissions globally have shifted ozone precursors into the tropics where ozone production is more efficient. Zhang et al. (2016) suggest that this equatorward shift of emissions has increased the total global ozone burden more than the combined effect of the increase in global methane emissions and the increase in the total mass of non-methane precursor emissions. Butler et al. (2020) however showed that surface ozone in the northern midlatitudes is more strongly influenced by emission sources in the northern midlatitudes than by tropical sources.

Contribution to Observed Trends

Global model simulations of annual average ozone have shown overall declines in Europe and North America since 1990, with overall increases in East and South Asia. In Europe, the contribution of both regional and extra-regional sources declined between 1990 and 2000. In North America, the contribution of regional sources declined, but extra-regional sources remained relatively constant. In both regions, the contribution of methane increased over time offsetting some of the benefits of regional emission control (Wild 2012).
Figure. Annual regional mean surface O3 changes relative to 2000 over each HTAP region following historical precursor emission changes between 1960 and 2000 (top row), and the contribution of regional anthropogenic sources, anthropogenic sources outside the region, and global methane changes (bottom row). Individual model responses are shown in grey and the mean of all 14 models is coloured. (From Figure 6, Wild 2012).

Between 1990 and 2014, in western North America, increases in Asian NOx emissions contributed an estimated 65% to the observed increase in springtime background ozone levels, while increases in global methane contributed an estimated 15%. In summer, increasing Asian emissions approximately offset the benefits of US emission reductions (Lin 2017).

For Europe, ground-level ozone concentrations are more sensitive to anthropogenic emissions outside of Europe than to anthropogenic emissions inside of Europe. This is the case for all European sub-regions, all seasons, and for a range of ozone metrics including annual and seasonal averages, SOMO35, and POD1 (Jonson 2018). The relative influence of these extra-regional emissions on ozone in Europe varies by location, season, and ozone metric. Extra-regional anthropogenic emissions of NOx and VOCs outside of Europe are estimated to contribute between 2-12 ppb of ozone depending on the season, with an annual average contribution of 4-8 ppb of ozone depending on the model used. The contribution of anthropogenic methane emissions to ozone in Europe is estimated to be about 5-8 ppb (on the basis of 6mDMA1).
For North America, region-wide annual average ozone concentrations appear equally sensitive to anthropogenic emissions outside of North America and emissions inside of North America. Extra-regional anthropogenic emissions of NO\textsubscript{x} and VOCs outside of North America and changes in global methane, together, are estimated to contribute between 8-13 ppb of ozone in the western United States, 2-12 ppb of ozone in the central United States, and 2-10 ppb of ozone in the eastern United States, depending on the season.

Although ozone concentrations are more sensitive to extra-regional sources than are PM concentrations, PM concentrations have a larger impact on mortality. Considering PM\textsubscript{2.5} and O\textsubscript{3} mortality effects together in regional models for Europe and North America, Im et al. (2018) found that a 20% decrease in global anthropogenic emissions of non-methane precursors of ozone and PM would result in a decrease of 54,000 and 27,500 premature deaths in Europe and North America, respectively. In Europe, 87% of the reduced deaths were associated with emission decreases within Europe, 2% were associated with decreases in North America, and 11% were associated with decreases in the rest of the world. In North America, 90% of the reduced deaths were associated with emission decreases in North America, with the remaining 10% due to emission decreases elsewhere.

Liang et al. (2018) found similar results to Im et al (2018) using an ensemble of global models. In contrast to the findings in HTAP (2010), Liang et al. (2018) found that emission decreases in North American and Europe resulted in more avoided ozone related deaths inside those regions than outside those regions. However, Europe, as well as the regions of Russia and Belarus and the Middle East, had more avoided deaths due to decreases of extra-regional emissions of O\textsubscript{3} precursors than decreases in regional emissions.

**Contribution to Projected Trends**

Turnock et al. (2018) found that annual average surface ozone concentrations in 2050 in Europe and North America are expected to be relatively similar to 2010 under a current legislation scenario, despite strong decreases in regional NO\textsubscript{x} emissions. In both Europe and North America, the contribution of extra-regional sources of non-methane precursors does not change much, but an expected increase in global methane concentrations offsets the decreases in regional emissions. Under a climate policy scenario, the increase due to methane emissions is not as high, North America benefits from decreased regional contributions, and Europe benefits from decreased extra-regional contributions. Under a maximum technically feasible scenario, the contributions of regional, extra-regional, and methane sources are all decreased, with the largest decreases coming from the control of extra-regional sources.
Europe

North America

Figure. Total annual mean change in regional surface O3 concentrations over Europe (left) and North America (right) and the contribution of local (blue), remote (red) and methane (gold) sources between 2010 and 2050 from the parameterisation for the ECLIPSE V5a emissions under the CLE (a), CLIM (b) and MTFR (c) scenarios. Grey lines on the local and methane panels represent individual model estimates of O3 changes, showing the spread in model responses; solid lines show the multi-model mean. Error bars represent 1 standard deviation over the model range. The last row of panels shows the O3 response from individual sources plotted together for each year. (Figures 7 and S6 of Turnock, 2018).

Turnock et al. (2020) describe the changes in ground-level ozone throughout the 21st century from the ensemble of CMIP6 models, based on emissions from SSPs 1, 2, 3, and 5. Surface ozone decreases in the UNECE region under SSPs 1 and 2, and either increases or stays roughly constant (depending on the world region) under SSPs 3 and 5. Short-lived ozone precursors (NOx and NMVOC) generally decrease in SSP5 and increase in SSP3. Increasing ozone in the SSPs is thus linked to increasing methane. A similar result was seen in the CMIP5 model ensemble using the RCP scenarios, in which short-lived ozone precursors decreased in all scenarios, but surface ozone increased in RCP8.5, the only scenario in which methane increased (Young et al. 2013).

Globally, health benefits from reduced PM burdens are projected to outweigh those from reduced ozone globally by more than a factor of 10, while ozone damage to crops is expected to result in approximately 10% or less additional production loss by 2030/2050 under high emissions (RCP8.5/6.0) scenarios (von Schneidemesser et al., 2020). Reduced emissions (RCP4.5, 2°C target) scenarios can lead to small global crop production gains of a few per cent. Changes to future production are expected to be larger in several regions, including India and China, but these are also more widely varied due to uncertain emissions trajectories.
References


Young et al. (2013) https://doi.org/10.5194/acp-13-2063-2013


Topic 2: Projected trends in methane, contribution to ground-level ozone, and mitigation potential

The one question under this topic is assigned to the hemispheric transport chapter of the GP review.

Questions under this topic

Question 3.3: What is the projected future trend in methane emissions? What is the impact on ozone formation? In which regions and in which sectors outside the UNECE region is there potential for emission reductions that have a significant effect on reducing ozone effects in the UNECE region?

This question was assigned to TF HTAP and MSC-W, however input from TFIAM will be essential to providing an answer.

Summary of the TF-HTAP contribution to this topic

8. Projected trends in anthropogenic methane emissions span a very wide range, between a factor of two smaller or a factor of two larger than present-day emissions by the end of the century, depending on assumptions made about economic development and the use of emission control technology.

9. Ozone formation is strongly influenced by the atmospheric methane burden, with model studies consistently showing that higher mixing ratios of methane lead to higher background mixing ratios of ground-level ozone.

10. Due to the long lifetime of methane in the atmosphere, methane is well mixed. Decreases in surface ozone arising from methane emission control are largely independent of source location, but the local response to global methane reduction is stronger in locations where local NOx emissions are high. Equal emission reductions in any given regions will lead to the same reductions in global background ground-level ozone.

11. The fossil fuel (production and distribution) and waste sectors have the highest technical potential for reduction of methane emissions. The agricultural sector is a major source of methane emissions but has a low technical potential for reductions in methane emissions.

12. Outside the UNECE region there is currently potential for reducing methane emissions from the waste sector in China and the fossil fuel sector in the Middle East.

Further explanation of the TF-HTAP contribution to this topic

Methane concentrations have been increasing at a rate of about 6 ppb per year in 2007-2013 and accelerating to 10 ppb per year during 2014-2018 (Dlugokencky 2020).

Approximately half of global methane emissions are anthropogenic, with the anthropogenic methane emissions being generally better constrained than natural emissions by both bottom-
up and top-down methods (Saunois et al. 2020). Projected trends in anthropogenic emissions of methane have been provided for the CMIP5 and CMIP6 exercises in the form of the RCP (Representative Concentration Pathways) and SSP (Shared Socioeconomic Pathways) scenarios respectively. The SSPs are described and compared with the earlier RCPs by Gidden et al. (2019). Figure X shows the projected emissions of methane for the RCP and SSP scenarios until 2100. Figure X shows the ECLIPSE5a scenarios developed by IIASA in comparison with the RCP and SSP scenarios until 2050.

Figure X: Methane emissions projections from the RCP and SSP scenarios until 2100 (Gidden et al. 2019, Figure3)

Figure 3.12. Historical (1990 – 2010) global anthropogenic CH4 emission trends from EDGAR v4.3.2 and projected (2000 – 2050) trends from four scenario families. Scenarios have been colour-coded to easily distinguish the "high emission", "middle of the road" and "low emission-high mitigation effort" members in each family.

Figure X: methane emissions projections from the ECLIPSE5a scenarios until 2050 compared with the RCP and SSP scenarios (van Dingenen et al., 2018, Figure 3.12)
There is no single projected future trend in methane emissions; the RCP and SSP scenarios span a wide range, depending on assumptions of socioeconomic development and mitigation levels. Methane emissions decrease (compared with the present day) in baseline versions of SSPs 1 and 2, and increase in baseline versions SSPs 3, 4, and 5. In the earlier RCP scenarios, methane emissions increase in RCP 8.5 and decrease or stay roughly constant in all three other scenarios. The ECLIPSE scenario “Current Legislation” falls roughly in the middle of the RCP and SSP “high emission” scenario set. The ECLIPSE “Climate” scenario, similarly to other “middle of the road” scenarios, shows a stabilization of methane emissions by 2050. The ECLIPSE “Maximum Technical Feasible Reduction” scenario shows a lower mitigation potential for global methane emissions than the comparable “high mitigation effort” set of RCP and SSP scenarios. Since the ECLISPE scenarios do not span as large a range as the RCP and SSP scenarios, the potential for mitigation of methane emissions (the difference between the highest emission and lowest emission scenarios) is lower in ECLIPSE. Most of this difference in mitigation potential between ECLIPSE and the RCP/SSP scenarios is seen in the Middle East + Africa and Asian regions (Figure below).

Figure X: Methane mitigation potential by 2050 from ECLIPSE scenarios compared with RCP and SSP (and GECO2017) scenarios (van Dingenen et al., 2018).

Trends in projected emissions from the energy, waste, and agricultural sectors dominate the projected future trends in methane emissions (eg. Figure below). Scenarios in which methane emissions decrease can be characterised either by a phasing out of reliance on fossil fuels or stronger mitigation measures. Scenarios in which methane emissions increase are characterised by increasing unmitigated emissions primarily from the energy and/or agricultural sectors. While emissions from the agricultural sector are high, the technical potential for emission reduction is low. In future scenarios under which methane emissions are successfully mitigated, the agriculture sector becomes a dominant source of anthropogenic methane emissions. Further mitigation of methane from the agricultural sector will require the use of structural measures such as replacing intensive feedlot farming with extensive grazing, and the modification of human diets. Scenarios representing the effects of these structural measures on methane and associated emissions are currently not well developed.
Butler et al. (2020) recently estimated that methane oxidation contributes to 40% of the annual average present-day northern hemisphere concentration of ground-level ozone. Turnock et al. (2020) describe the changes in ground-level ozone throughout the 21st century from the
ensemble of CMIP6 models, based on emissions from SSPs 1, 2, 3, and 5. Surface ozone decreases in the UNECE region under SSPs 1 and 2, and either increases or stays roughly constant (depending on the world region) under SSPs 3 and 5. Short-lived ozone precursors (NOx and NMVOC) generally decrease in SSP5 and increase in SSP3. Increasing ozone in the SSPs is thus linked to increasing methane. A similar result was seen in the CMIP5 model ensemble using the RCP scenarios, in which short-lived ozone precursors decreased in all scenarios, but surface ozone increased in RCP8.5, the only scenario in which methane increased (Young et al. 2013).

Fiore et al. (2008) showed that decreases in surface ozone arising from methane emission control are largely independent of source location, but the local response to global methane reduction is stronger in locations where local NOx emissions are high. In Europe, changes in emissions outside Europe and global methane concentrations will largely drive future annual average O3 levels. Without additional controls, global methane emissions are expected to grow, increasing O3 mortality in Europe in 2050 by up to 8,000 additional premature deaths compared to 2010 levels. Implementation of mitigation policies, largely outside of Europe, can decrease methane emissions overall and decrease O3 mortality in Europe by up to 2000 premature deaths per year compared to 2010 levels, a difference of 10,000 deaths per year between the highest and lowest global CH4 emissions scenarios. In North America, the difference between the highest and lowest global CH4 emissions scenarios corresponds to a difference of up to 5,000 deaths per year in 2050. The sectors with substantial mitigation potential are fossil fuel production, waste and wastewater management, and agriculture, with the largest emissions in China, followed by Latin America, Africa, India, and North America (vanDingenen 2018).

References


Young et al. (2013) [https://doi.org/10.5194/acp-13-2063-2013](https://doi.org/10.5194/acp-13-2063-2013)
Topic 3: Projected trends in international shipping, contribution to ground-level ozone and N deposition, and mitigation potential

The one question under this topic is assigned to the hemispheric transport chapter of the GP review.

Questions under this topic

Question 3.4: What is the projected future trend in NOx-emissions from shipping? What is the impact on ozone formation and nitrogen deposition? What and where is the potential for emission reductions that have a significant effect on reducing ozone effects in the UNECE region?

This question was assigned to TF HTAP and MSC-W. MSC-W and TFIAM have done recent work on the impacts of shipping on the EMEP region. TF HTAP can provide additional context from HTAP2 experiments and from the literature. TF HTAP is also performing new analyses that may provide additional insights about the role of shipping activity beyond coastal regions.

Summary of the TF-HTAP contribution to this topic

13. NOx emissions from international shipping on the global seas are projected to remain approximately constant or decrease slightly in absolute terms over the 21st century, depending on assumptions about growth in international trade and the use of emission control technology. The share of global shipping NOx as a proportion of global anthropogenic NOx emissions (currently at about 30%) is projected to vary between 10% and 60%, by the end of the century depending on the effectiveness of land-based NOx emission control.

14. Projections of the future effects of shipping on air quality in Europe has focused on the human health impact of PM2.5 from SOx and NOx emissions over European seas. Projections of the impact of global shipping NOx on baseline ground level ozone and N deposition in the UNECE region are currently lacking. Models show low agreement on the present-day effects of shipping NOx on ground-level ozone, but do agree that extra-regional sources account for up to half of N deposition in coastal regions, strongly indicating a role for shipping NOx.

15. Due to the short lifetime of NOx, it seems likely that reduction of emissions of ship NOx near coastlines has a high potential to reduce N deposition. There are some indications that the global springtime maximum in intercontinental transport of ozone is influenced by shipping NOx emitted over the high seas, but further model studies are needed to determine the strength of this influence.
Further explanation of the TF-HTAP contribution to this topic

Globally, the absolute NOx emissions from shipping in the SSP scenarios tend to decrease in the 21st century, at a rate depending on the socioeconomic development pathway assumed (Rao et al. 2017). In SSPs 1 and 3, which are characterised by deglobalisation, NOx emissions decrease faster than in SSPs 2 and 5, which are characterised by increasing globalisation. Shipping NOx emissions as a proportion of total global NOx emissions decreases in SSPs 1 and 3, stays roughly constant in SSP5, and increases from ~30% to 60% by 2100 in SSP2, due to the combination of high shipping volumes and stronger reduction of land-based NOx emissions in SSP2 (Fig. X).

Figure X: Absolute emissions of NO from international shipping in the SSP scenarios (left) and ship NO as a percentage of total global emissions (right). Figures prepared by A. Nalam (IASS Potsdam) based on SSP scenario emissions as described by Rao et al. (2017)

We are not aware of any studies specifically examining the effects of ship NOx on ozone or N deposition using the SSP scenarios. Several studies have examined these effects in the present day or recent historical periods.

NOx emissions from shipping in European seas are projected to exceed European land-based sources by 2030 (Cofala et al., 2018). There is substantial scope to reduce shipping NOx emissions over European seas through both climate measures and additional technical emission control (Figure below). The effect of these NOx emission reductions on ground-level ozone and N deposition over Europe was not quantified by Cofala et al. (2018).
Butler et al. (2020) noted a strong contribution of ship NOx to the springtime maximum in long-range transboundary ozone in all regions of the northern hemisphere, and a strong contribution of ship NOx to European summer ozone, the latter most likely due to nearby NOx sources. Jonson et al. (2018) compared the predictions of several models for ozone in Europe and found a high degree of variability in the contribution of ship NOx across different models. Representation of chemistry and dilution in ship plumes is currently a weakness of global models (see the answer to Question 2.7).

We are not aware of any studies attributing ozone or N deposition to ship NOx from different world regions using models at the global or hemispheric scales. Butler et al. (2020) and Jonson et al. (2020) both suggest that NOx emissions over the high seas have a strong influence on European surface ozone, especially in the springtime.

The sensitivity of the deposition of sulfur (S), oxidized nitrogen (NOy), and reduced nitrogen (NHx) to changes in extra-regional anthropogenic emissions is similar to the sensitivity of PM$_{2.5}$ concentrations. Using the results of 11 global models, the RERER for S deposition in Europe was estimated to be 0.36 averaged regionally, with values of 0.53 for coastal areas and 0.27 for non-coastal areas. The RERER for NOy deposition in Europe is 0.34 (0.48 for coastal areas, 0.27 for non-coastal areas). For NHx deposition, the RERER across Europe in 0.12 (0.22 in coastal areas, 0.09 for non-coastal areas). In North America, the RERERs are smaller and show a similar pattern: for S deposition, 0.17 overall (0.40 in coastal areas, 0.12 in non-coastal areas); for NOy deposition, 0.17 overall (0.43 in coastal areas, 0.12 in non-coastal areas); and
for NH₃ deposition, 0.07 overall (0.31 in coastal areas, 0.05 in non-coastal areas). The larger
sensitivity in coastal areas may be due to the location of coastal areas in proximity to upwind
sources, including maritime shipping, and differences in deposition processes in coastal
regions.

In North America, 83% of S deposition, 83% of NOy deposition, and 93% of NH₃ deposition are
due to sources within North America. In Europe, the fractions are 64%, 66%, and 88% for S,
NOy, and NH₃ deposition, respectively. However, in the region defined by Russia, Ukraine, and
Belarus, only 39%, 41%, and 45% of S, NOy, and NH₃ deposition, respectively, are due to
emissions within the region. The percent of emissions transported and deposited outside each
of the priority source regions is shown in Table 2.

Table 2. Percent of emissions in a source region transported out of the region and
deposited elsewhere (Tan 2018).

<table>
<thead>
<tr>
<th>Source Region</th>
<th>S</th>
<th>NOy</th>
<th>NHx</th>
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<tbody>
<tr>
<td>North America</td>
<td>31%</td>
<td>29%</td>
<td>12%</td>
</tr>
<tr>
<td>Europe</td>
<td>40%</td>
<td>34%</td>
<td>17%</td>
</tr>
<tr>
<td>Russia/Belarus/Ukraine</td>
<td>38%</td>
<td>39%</td>
<td>23%</td>
</tr>
<tr>
<td>East Asia</td>
<td>27%</td>
<td>26%</td>
<td></td>
</tr>
<tr>
<td>South Asia</td>
<td>34%</td>
<td>34%</td>
<td>15%</td>
</tr>
<tr>
<td>Middle East</td>
<td>58%</td>
<td>46%</td>
<td>51%</td>
</tr>
</tbody>
</table>

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Topic 4: Sufficiency of atmospheric modelling for understanding hemispheric transport of air pollution, and the main requirements for improving simulation of hemispheric transport

Both questions under this topic are assigned to a chapter in the GP review devoted to the measurement and modelling activities of the convention. TF-HTAP will also cover the hemispheric aspects of this topic in the chapter on hemispheric transport.

Questions under this topic

Question 2.6: What has been the influence of improved atmospheric modelling (e.g. the higher spatial resolution) on the effectiveness of emission reductions for air quality improvement and deposition? Did this increase the challenge to meet environmental quality and health targets?

This question was assigned to MSC-W and TF HTAP. MSC-W is best equipped to comment on how improvements in the EMEP model have changed estimates of source-receptor relationships and the estimated effectiveness of emission reductions at the regional scale. TF HTAP should focus on how model improvements have changed estimates of intercontinental transport and the contribution of extra-regional emissions sources to impacts within the UNECE region.

Question 2.7: Is the monitoring and modelling system of the Convention sufficient to observe, assess and project air pollution and its effects related to the Gothenburg Protocol in the UNECE region? If not, what are the main challenges and what is needed to meet them?

This question was assigned to all of the bodies under WGE and EMEP. TF HTAP should focus on the sufficiency of available models, emissions inventories, and observational data at the global scale to provide estimates of the impact of extra-regional emission sources on impacts in the UNECE region.

Summary of the TF-HTAP contribution to this topic

16. Multi-model intercomparisons show a very large spread in simulated surface ozone, which has not improved over the last decade despite higher spatial resolution and other model developments. As an ensemble, global models tend to overestimate available surface observations.

17. The source/receptor relationships for ground-level ozone from the HTAP2 multi-model exercise were not significantly different from those of the HTAP1 exercise, despite developments in individual models and closer harmonisation of the model inputs.

18. Global models disagree strongly on the magnitude of the pre-industrial to present-day trend in ground-level ozone, and tend to underestimate the magnitude of the observed trend.
Projection of the contribution of hemispheric background ozone to the attainment of future targets using current models remains highly uncertain.

19. Regional ozone models generally performed better in comparison to observations than did global ozone models, which generally have lower spatial resolution than regional models. However, the best performing global models compared better to observations than did the worst performing regional models.

20. Technical challenges for improved global simulations of ground-level ozone for the UNECE region include more accurate simulation of the global methane lifetime, better resolution of the NOx chemistry of ship exhaust plumes, and better representation of ozone deposition to vegetation.

21. Model intercomparison studies such as HTAP, CCMI, and AerChemMIP exercises play a vital role in assessing the adequacy of state-of-the-art emission inventories, global models, and measurement data for informing the Convention on the impacts of extra-regional emission sources on ozone impacts in the UNECE region.

22. In addition to model development, ongoing provision of high-quality emission inventories and expansion of the global network of ozone observations for model evaluation are required.

Further explanation of the TF-HTAP contribution to this topic

Sufficiency of atmospheric models for understanding hemispheric transport

In HTAP2, the differences in predictions of particulate matter concentrations from different models decreased relative to the HTAP1 but this was not the case for ozone concentrations. The lack of improvement in model agreement for ozone was surprising given that all HTAP2 models used the same emissions inputs, whereas in HTAP1 emissions inputs were not harmonized. More work is needed to understand the processes contributing to the spread in model predictions, but analysis to date suggests that differences in the vertical transport and deposition processes between models may be resulting in this spread. Higher resolution regional models generally performed better in comparison to observations than did the coarser resolution global models. However, the best performing global models compared better to observations than did the worst performing regional models (Solazzo 2016, Solazzo 2017).

A large inter-model spread in modelled surface ozone was also noted by Young et al. (2013) from the ACCMIP study (their Fig. 3). More recently, Turnock et al. (2020) also noted a large spread in surface ozone from models contributing to CMIP6 (their Fig. 4, reproduced below as Fig. X). The global models involved in these studies show better agreement on free tropospheric ozone than ground-level ozone, which is more difficult to simulate due to the influence of additional processes such as vertical mixing in the Planetary Boundary Layer and ozone dry deposition to vegetation.

Parrish et al. (2014) provides a detailed comparison of observed and modelled long-term trends for the northern midlatitudes. Models tend to underestimate the long-term (multi-decadal) changes in surface ozone. Turnock et al. (2020) showed that the CMIP6 models disagree
strongly on the modelled pre-industrial to present-day trend of surface ozone (their Fig. 9 reproduced below as Fig. X).

Lin et al (2015) found that sampling frequency and distribution of ozone profile observations may account for some of the differences between observed and model estimates of ozone trends in western North America. When corrected for the uncertainty in data representativeness, the observational trend estimate and modeled trend estimate overlapped.

**Figure X:** Figure 4 from Turnock et al. (2020) showing CMIP6 model ensemble comparison with TOAR observations.
Figure 9 - Changes in the regional and global annual mean surface O₃ concentrations, relative to a 2005-2014 mean value, across 5 CMIP6 models and the HTAP_param. The multi-model annual mean year 2005-2014 surface O₃ concentrations (+/- 1 standard deviation) are shown in the top left of each panel. Regions are defined in Figure S1.

Figure X: Figure 9 from Turnock et al. (2020) showing the spread of CMIP6 model estimates for the pre-industrial to present-day trend in ground-level ozone.

Main requirements for improved simulation of hemispheric transport

Increased model resolution can improve the agreement between models and observations of surface ozone, although it is not always clear whether the better agreement is due to improved model process representation, or improved model representation of the spatial scales represented by the measurements. Models can simulate European rural background ozone sufficiently with ~15x15km resolution (Schaap et al. 2015). Regional chemical transport models for Europe are commonly run at this resolution or even higher. Regional models may perform better than global models for Europe due to their higher spatial resolution, but also due to their being more highly constrained than global models. Simulation of intercontinental transport requires the use of global models, which are not commonly run at resolutions finer than the order of about 100km resolution. In the HTAP1 multi-model exercise, models were run with resolutions of about 200x200km, and in the subsequent HTAP2 exercise, the finest resolution
for a global model increased to about 50x50km. There is still scope for improvement in the spatial resolution of global models for the simulation of background ozone.

Given the possible importance of NOx emissions from shipping for long-range transport of ozone into the UNECE region in springtime (see Topic 3), global models require improved treatment of the chemistry of ship exhaust plumes to sufficiently assess and project the impact of ship NOx emissions. The relatively coarse resolution of global models is not sufficient to resolve the small scale of ship exhaust plumes (Kasibhatla, 2000, von Glasow 2003). In effect, ship NOx emissions in global models are instantaneously diluted into large grid cells which are otherwise relatively low in NOx. This increases the modelled lifetime of NOx and the ozone production over the oceans, leading to overestimation of the effect of ship NOx on ozone production. Approaches have been developed to account for the high-NOx chemistry of ship exhaust plumes in the otherwise clean marine boundary layer (eg. Vinken et al. 2011), but these have not been widely implemented in current generation global models.

Given the importance of methane oxidation as a precursor for ground-level ozone (see Topic 2), the large spread in modelled tropospheric methane lifetimes in the current generation of global models is another challenge. This spread has been around 7-10 years in several recent model intercomparison exercises (Saunois et al. 2020). Such a spread in modelled methane oxidation rates could be expected to lead to a large uncertainty in modelled ozone production due to methane oxidation in these models, but no model intercomparison exercise has yet systematically quantified this uncertainty. The current practice of forcing global model simulations using prescribed methane concentrations should also be revisited. The chemical sink of methane is influenced by both future emissions and climate change. Model experiments driven by prescribed concentrations do not take this into account.

The mechanistic representation of the multiple pathways of ozone deposition and associated atmosphere-vegetation feedbacks is currently highly simplified in current global models (eg. Sadiq et al. 2017, Lin et al. 2019, Clifton et al. 2020). Ozone deposition to vegetation is also a major cause of damage to crops and ecosystems (Emberson et al. 2013). Improvement of the representation of ozone deposition processes in models will lead to improved estimates of ozone impacts on both vegetation (through improved deposition flux estimates) and human health (through improved concentration and exposure estimates).

Model intercomparison exercises are an important method for understanding the uncertainties in state-of-the-art models, and thus their limitations for assessment and projection of air pollution and its effects. Such exercises characterize the ranges of differences between models, and can have the potential to reveal deficiencies in models process representations and point the way towards improvements. A regular cycle of model intercomparison exercises allows the tracking of improvements in models and how these translate into better assessment and projection of air pollution and its effects. Intercomparison exercises may be ongoing exercises with a focus on process representation and model evaluation (eg. CCMI, Duncan et al. 2016), may focus on chemistry-climate interactions and periodically feed into IPCC assessments (eg. Lamarque et al. 2013, Collins et al. 2017), or may be specifically designed to quantify intercontinental transport of air pollution and feed into the processes and timelines of the LRTAP Convention (HTAP
Ongoing and coordinated model intercomparison exercises are vital for continuous assessment of the sufficiency of state-of-the-art models for informing the LRTAP convention on air pollution and its effects related to the Gothenburg Protocol in the UNECE region.

Accurate emission inventories are vital for accurate model assessment of air pollution and its impacts. For assessment of intercontinental transport of ozone, emission inventories must be accurate both within and outside the UNECE region. Mosaic inventories such as the EDGAR-HTAPv2 inventory (Janssens-Maenhout 2015) combine detailed regional inventories in a consistent global framework and are especially useful for assessment and projection of intercontinental transport of air pollution.

Global models do not currently produce all features of the observed distribution and trends in ground-level ozone (see the answer to Question 2.6). Ongoing efforts to collect, characterize, synthesise, and distribute ozone observations and associated metrics for model evaluation both within and outside the UNECE region such as the TOAR project (Schultz et al. 2017) are vital for continued evaluation and improvement of models.

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